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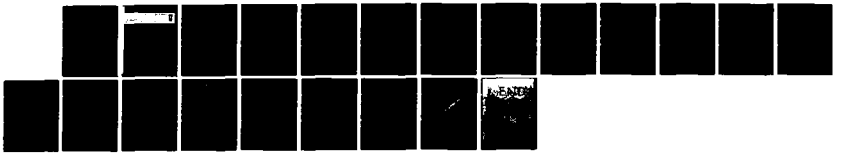

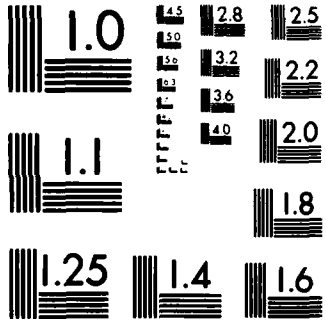

MICROCOPY RESOLUTION TEST CHART
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TECHNION Israel Institute of Technology
Department of Aeronautical Engineering

March 1983

TAE No. 503

Ignition Delay Times of Propene Oxygen Argon Mixtures

by

Alexander Burcat and Josef Levitas

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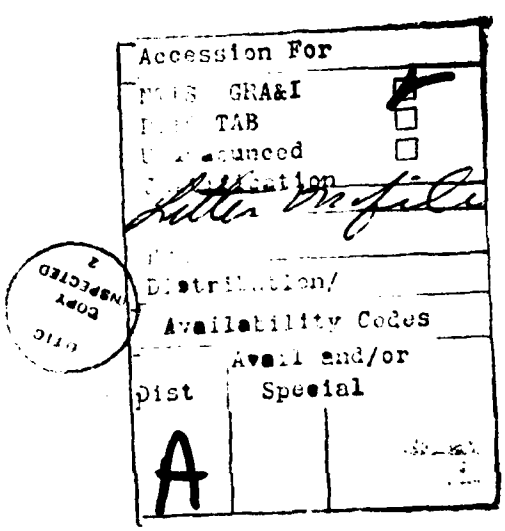

ABSTRACT

The ignition delay times of Propene Oxygen Argon mixtures were measured in a shock tube. The concentrations ranged from 0.6% to 1.6% C_3H_6 and 2.7%to 14.4% oxygen. The initial pressures were 50 to 133 torr, and the temperatures achieved were 1274 -1848 K.

The overall ignition delay equation for Propene Oxygen Argon mixtures has a value of

$\tau = 5.3 \times 10^{-14} \exp[(37.5 \times 10^3)/RT][C_3H_6]^{0.23} [O_2]^{-1.12} [Ar]^0$ sec

A kinetic scheme for the oxidation of propene is proposed.

INTRODUCTION

Propene is an important raw material in the production of polypropylene plastics. It was also found to be an intermediate in the decomposition of propane [1], the first step to occur by spontaneous thermal dehydrogenation. The decomposition was also found to occur in considerable quantities before any oxidation starts in the ignition process of propane [2].

When measuring the ignition delay time of propane [2,3], it is completely unclear whether the correlation found is that of propane or that of propane + propene + propyne, the latter being a degradation product of propene itself [4].

Thus the ignition delay values of propene are very important in elucidating this question.

EXPERIMENTAL

The ignition experiments of propene were performed in a stainless steel shock tube whose diagram is given in Fig. 1. The tube is 50 mm in diameter (two inches) and 4 meters long. The driven section is 2.5 meters long. Mylar diaphragms of different gauges are used. They are burst by the driver's gas pressure. The driver gas used in this study was Airco pure grade helium. Two specially built valves divide the driven section. One of the valves is located in the middle of the tube thus permitting half of the tube to be filled with the reacting mixture while the other half is filled with a matched-impedance neutral mixture or argon.

The second valve allows separation of the end block of the shock tube from the main instrument in order to extract gas samples from the tube for analysis. The shock speed is measured near the end block over a distance of 200 mm by two piezoelectric gauges connected to a HP 3400 counter with a

resolution of ± 0.1 μsec. The pressure is measured by a Kistler 603A piezoelectric gauge mounted into the end plate. A home made thin platinum heat transfer gauge is also mounted near the end plate of the shock tube.

The gases used were Matheson Propylene C.P. 99.0% pure, I.B. Miller Oxygen 99.5% pure and 99.9% pure argon.

Gas mixtures were prepared manometrically in stainless steel cylinders using a high pressure manifold. They were left overnight to mix before use. All mixtures used are listed in Table I.

The reflected shock temperatures were calculated using the standard conservation equations and the ideal gas equation of state, assuming frozen chemistry. The enthalpies of propylene were recalculated at the proper temperature range using statistical methods [5] and well determined spectroscopic data [6]. They were published elsewhere [7]. Oxygen and argon thermodynamic properties were taken from thermodynamic tables [8,9].

RESULTS AND DISCUSSION

As shown in Table I 6 propylene - oxygen - argon mixtures were used for the seven series of experiments for a total of 118 shocks. The mixtures were prepared in such a way as to permit direct deliniation of the power dependencies of the reactants in the assumed ignition delay equation

$$\tau = 10^{-x} \exp(+E/RT) [C_3H_6]^a [O_2]^b [Ar]^c \text{ sec}$$

In each of the performed shocks, the exact mixture and initial pressure were known. The recorded post shock experimental properties were the reflected temperature T_5, the density ratio ρ_1/ρ_5 and the ignition delay time τ. The experiments were spread over the temperature range in such a way as to permit maximum sensitivity to the determination of the so called "activation energy".

Figure 2 represents series A, B and C in a log τ vs $1/T_5$ graph. Mixtures B and C have a threefold difference in the argon concentration (normalized for density ratio differences). The experiments show a large scatter and a mixture of the data of the two series, suggesting negligible or very small argon power dependency.

Figure 3 represents the log τ vs $1/T_5$ for series D and E. The distance among the two lines is very small, approximately 0.08 log τ units. Therefore, the propene power dependency should be:

$$\log \tau_E = a \log (0.8\% C_3H_6) + b \log (7.2\% O_2) + c \log A$$

$$\log \tau_D = a \log (4 \times 0.8\% C_3H_6) + b \log (7.2\% O_2) + c \log A$$

substracting τ_E from τ_D and disregarding the small differences in argon concentration

$$\log (\tau_E - \tau_D) = a \log 4$$

$$a = \frac{\log (\tau_E - \tau_D)}{\log 4} = \frac{0.08}{0.6} = 0.138$$

Figure 4 represents groups F and G on a logτ vs $1/T_5$ graph. As explained above the oxygen dependency can also be evaluated from the graph and it is found that b = -1.33.

Figure 5 represents the overall plot of log β vs 1/T for a maximum acceptable spread of 2σ. The correlation was found with a statistical "student-t" program

$$\tau = 5.3 \times 10^{-14} \exp(+(37150 \pm 1860)/RT)[C_3H_6]^{0.15 \pm 0.08}[O_2]^{-1.12 \pm 0.09}[Ar]^{0.20 \pm 0.198} \text{ sec}$$

This correlation is found with 103 shocks. Using a 3σ correlation with 115 shocks the data found are

$$\tau = 1.9 \times 10^{-14} \exp(+(37680 \pm 2326)/RT)[C_3H_6]^{0.23 \pm 0.1}[O_2]^{-1.11 \pm 0.12}[Ar]^{0.0} \text{ sec}$$

Table II contains a list of all shocks performed in this study and their pre and post shock parameters.

This investigation should be regarded in the proper perspective and compared with earlier studies on propane and propyne ignition. Burcat et al. [2] have reported the following equation for propane

$$\tau = 4.4 \times 10^{-14} \exp[(42.2\times10^{3})/RT][C_3H_8]^{0.57}[O_2]^{-1.22}[Ar]^{0} \text{ sec}$$

The findings of this work on propene gives the following value

$$\tau = 5.3 \times 10^{-14} \exp[(37.5\times10^{-3})/RT][C_3H_6]^{0.23}[O_2]^{-1.12}[Ar]^{0} \text{ sec}$$

Unpublished results of propyne ignition [10] read approximately

$$\tau = 4 \times 10^{-12} \exp[(27.8\times10^{-3})/RT][C_3H_4]^{-0.04}[O_2]^{-0.92}[Ar]^{0} \text{ sec}$$

Therefore, the reasoning should be as follows: Earlier studies [1,2] have shown that propene is easily formed during the propane decomposition and that extensive decomposition occurs before oxidation steps start to influence.

The big similarity in ignition delay values of propane and propene suggest that the values measured for propane are really those of a mixture that contains mainly propylene and the propene influence is very great. However, propyne's influence on propane and propene is smaller.

The kinetic scheme of propene ignition should be calculated using earlier propane oxidation schemes [3] and eliminating from them the propane reactions such as the first 13 steps as proposed by Jachimowski [3]. In their place some of the elementary steps proposed recently by Kiefer [11] should be included.

The proposed scheme should include a list of reactions from different sources [3, 10-14]. Table III lists all the proposed scheme and the rate constants found in the literature.

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14) G.S. Bahn, "Theoretical Nitric Oxide Production Incidental to Autoignition and Combustion of Several Fuels", NASA CR-2455 (1975).

Table I. Experimental conditions of propylene mixtures.

Series	Concentrations % C_3H_6	O_2	Ar	P_1 ~ torr	Number of experiments	Parameters detected
A	1.6	7.2	91.2	100	16	-
B	1.6	7.2	91.2	50	13	c
C	0.6	7.2	96.7	133	17	c
D	3.2	7.2	89.6	100	17	a
E	0.8	7.2	92.0	100	20	a
F	1.6	14.4	84.0	100	21	b
G	1.6	3.6	94.8	100	14	b

TABLE II

INCIDENT AND REFLECTED SHOCK PARAMETERS FOR A MIXTURE OF 3 SUBSTANCES.

THE MOLE FRACTION OF C3H6 IS 0.01600

A THE MOLE FRACTION OF O2 IS 0.07200

THE MOLE FRACTION OF AR IS 0.91200

NO	P1	UPIOT	TREF	UREF	TAU	PS(ATM)
1	105.	6.78	1420.8	0.4172	517.0	4.4333
2	90.	6.71	1402.0	0.4153	629.0	3.9631
3	87.	6.62	1375.1	0.4125	842.0	3.8749
4	85.	7.36	1592.2	0.4340	122.0	4.3650
5	94.	7.61	1704.2	0.4459	70.0	5.6333
6	97.	6.86	1446.2	0.4199	385.0	4.2192
7	106.	7.50	1663.3	0.4410	61.0	5.4730
8	90.	7.04	1505.8	0.4261	213.0	4.5594
9	91.	7.58	1690.8	0.4446	61.0	5.1133
10	87.	7.67	1728.5	0.4479	61.0	5.0452
11	90.	7.28	1586.2	0.4343	77.0	4.9062
12	97.	7.10	1545.2	0.4301	133.0	4.7092
13	97.	7.12	1530.3	0.4286	113.0	4.6351
14	112.	7.34	1606.8	0.4363	113.0	5.7959
15	97.	7.00	1491.7	0.4246	307.2	4.4425
16	102.	6.59	1366.0	0.4119	650.0	4.0255

THE MOLE FRACTION OF C3H6 IS 0.01600

B THE MOLE FRACTION OF O2 IS 0.07200

THE MOLE FRACTION OF AR IS 0.91200

NO	P1	UPIOT	TREF	UREF	TAU	PS(ATM)
17	51.	7.54	1670.0	0.4435	66.6	2.8334
20	47.	7.30	1592.2	0.4349	206.0	2.6065
21	49.	7.30	1593.4	0.4350	215.0	2.5015
23	50.	6.86	1452.4	0.4205	922.0	2.1905
24	54.	6.63	1376.9	0.4127	1331.0	2.1020
25	49.	7.53	1672.8	0.4428	122.9	2.7054
26	49.	7.06	1518.0	0.4273	512.0	2.3105
27	51.	7.00	1491.7	0.4246	532.5	2.3356
28	51.	7.18	1549.8	0.4306	389.0	2.4680
29	51.	7.05	1509.1	0.4264	496.3	2.3614
30	54.	7.33	1603.1	0.4359	308.0	2.7641

THE MOLE FRACTION OF C3H6 IS 0.00600

C THE MOLE FRACTION OF O2 IS 0.02700

THE MOLE FRACTION OF AR IS 0.96700

NO	P1	UPIOT	TREF	UREF	TAU	PS(ATM)
101	140.	6.37	1626.5	0.4612	235.5	6.3631
102	173.	6.65	1761.7	0.4758	61.4	8.8356
103	133.	6.59	1728.3	0.4722	102.4	6.4589
104	131.	6.52	1695.0	0.4688	71.7	6.3519
105	134.	6.22	1559.2	0.4537	563.0	5.7005
106	136.	6.12	1518.3	0.4492	583.7	5.5466
107	138.	6.30	1593.5	0.4574	296.7	6.0744
108	138.	6.32	1601.9	0.4585	184.3	6.1251
109	135.	6.17	1547.4	0.4513	481.5	5.6162
110	135.	6.19	1543.1	0.4510	583.7	5.6405
113	130.	6.07	1830.1	0.4850	91.0	7.0785
115	130.	6.58	1725.6	0.4719	61.4	6.8734
117	129.	6.17	1550.7	0.4516	486.4	5.3702
118	133.	6.53	1699.0	0.4692	91.9	6.4724
119	138.	6.28	1583.0	0.4565	460.6	6.0173
120	138.	6.33	1608.0	0.4592	432.1	6.1614
121	135.	6.47	1670.0	0.4660	163.6	6.3980

THE MOLE FRACTION OF C3H8 IS 0.03200

THE MOLE FRACTION OF O2 IS 0.07200

THE MOLE FRACTION OF AR IS 0.89600

NO	P1	DPTOT	TREF	UPLE	TAU	PE(ATM)
302	99.	7.85	1443.7	0.3996	737.3	4.9237
303	97.	8.26	1547.1	0.4093	215.0	5.4347
304	99.	8.06	1495.6	0.4045	337.9	5.2341
305	99.	8.21	1533.4	0.4060	235.5	5.0632
306	100.	8.16	1522.1	0.4060	245.8	5.0490
307	99.	8.15	1519.9	0.4067	311.3	5.3611
308	100.	7.79	1428.4	0.3981	645.1	4.8622
309	100.	8.12	1511.0	0.4059	276.5	5.3607
310	99.	8.16	1521.0	0.4060	297.0	5.3876
311	100.	8.77	1687.0	0.4220	81.9	6.4891
312	103.	8.05	1493.3	0.4042	400.3	5.4251
313	100.	8.89	1722.8	0.4250	41.0	6.7145
314	102.	8.85	1712.0	0.4241	61.4	6.7766
315	101.	9.01	1750.0	0.4282	30.7	7.0185
316	99.	8.14	1515.8	0.4063	465.9	5.3539
317	103.	8.44	1596.7	0.4138	143.4	6.0688
318	99.	8.52	1618.8	0.4158	143.4	5.9895

THE MOLE FRACTION OF C3H8 IS 0.00800

THE MOLE FRACTION OF O2 IS 0.07200

THE MOLE FRACTION OF AR IS 0.92000

NO	P1	DPTOT	TREF	UPLE	TAU	PE(ATM)
401	100.	6.46	1453.3	0.4323	604.0	4.0805
402	103.	7.01	1688.3	0.4573	72.0	5.3432
403	100.	6.93	1655.6	0.4539	123.0	5.0310
404	100.	6.80	1603.1	0.4484	123.0	4.7603
405	100.	7.04	1703.0	0.4588	72.0	5.2584
406	101.	6.95	1664.7	0.4548	113.0	5.1250
407	97.	7.30	1772.8	0.4659	31.0	5.8274
408	102.	6.84	1620.3	0.4502	102.0	4.9596
409	100.	6.91	1646.8	0.4530	65.5	4.9080
410	99.	6.57	1515.7	0.4391	235.0	4.3257
411	99.	6.25	1400.9	0.4265	532.0	3.8023
412	102.	6.72	1571.3	0.4451	164.0	4.7254
413	100.	6.81	1456.5	0.4327	409.6	4.0948
414	100.	6.80	1454.4	0.4324	379.0	4.0652
415	101.	6.34	1432.8	0.4301	532.0	4.0259
416	100.	6.22	1392.2	0.4256	901.6	3.8009
417	99.	6.33	1429.8	0.4297	573.0	3.9326
418	100.	6.75	1574.3	0.4454	174.1	4.6439
419	101.	6.44	1465.9	0.4337	471.0	4.1797

THE MOLE FRACTION OF C3H6 IS 0.01600

THE MOLE FRACTION OF O2 IS 0.14400

THE MOLE FRACTION OF AR IS 0.84000

NO	P1	DRTOT	TREF	URLF	TAU	PE(ATM)
201	95.	7.83	1853.7	0.4129	117.0	4.5019
202	99.	7.51	1874.9	0.4150	136.5	4.8020
203	102.	7.68	1525.5	0.4209	117.0	5.2437
204	100.	7.45	1858.9	0.4138	273.0	4.7681
205	99.	7.40	1844.3	0.4120	253.5	4.6402
206	92.	7.35	1820.0	0.4105	370.5	4.2303
207	94.	7.62	1507.6	0.4183	136.5	4.8601
208	96.	7.35	1820.0	0.4105	331.5	4.5158
209	99.	7.16	1380.5	0.4055	409.0	4.2945
210	100.	7.82	1849.5	0.4124	204.8	4.7159
211	101.	7.48	1866.3	0.4142	189.4	4.8570
212	100.	7.01	1341.1	0.4015	614.4	4.3732
213	103.	7.85	1569.7	0.4248	76.8	5.5522
214	101.	7.64	1512.1	0.4187	128.0	5.1153
215	102.	8.06	1647.6	0.4318	30.9	5.0552
216	97.	8.05	1631.0	0.4302	71.7	5.5701
217	102.	7.73	1550.3	0.4213	92.2	5.3225
218	100.	7.38	1439.1	0.4114	225.3	4.6587
220	98.	7.96	1611.0	0.4263	61.4	5.4016
221	100.	6.74	1272.9	0.3945	891.0	3.8765
		6.98	1331.1	0.4004	594.0	4.0732

THE MOLE FRACTION OF C3H6 IS 0.01600

THE MOLE FRACTION OF O2 IS 0.07200

THE MOLE FRACTION OF AR IS 0.94800

NO	P1	DRTOT	TREF	URLF	TAU	PE(ATM)
502	100.	7.29	1700.7	0.4505	174.0	5.4599
503	100.	7.53	1799.6	0.4602	92.2	5.9463
504	100.	7.68	1863.5	0.4663	45.1	6.2729
505	99.	7.85	1936.9	0.4732	77.9	6.5987
506	99.	7.12	1633.6	0.4437	287.0	5.0403
507	97.	6.97	1578.9	0.4382	309.0	4.7791
508	97.	7.34	1710.5	0.4523	133.0	5.3687
509	103.	6.91	1556.6	0.4359	553.0	4.9500
510	97.	6.82	1523.6	0.4325	737.0	4.4187
511	103.	7.06	1611.4	0.4415	307.0	5.1385
512	104.	7.66	1853.7	0.4654	87.0	6.2289
513	99.	7.12	1634.9	0.4439	184.0	5.0554
514	98.	7.29	1698.9	0.4502	137.0	5.3170

Table III. Kinetic Scheme for Propene Oxidation

		A	12	E $\frac{kcal}{mole}$	Ref
1)	$C_3H_6 + M \rightleftharpoons CH_3 + C_2H_3 + M$	5.4×10^{75}	-15.71	120	11
2)	$C_3H_6 + H \rightleftharpoons C_3H_4 + H_2 + H$	1.6×10^{13}		0	11
3)	$C_3H_6 + CH_3 \rightleftharpoons CH_4 + C_3H_4 + H$	2×10^{13}		14.0	13
4)	$iC_3H_7 \rightleftharpoons C_3H_6 + H$	2×10^{14}		41.3	11
5)	$iC_3H_7 \rightleftharpoons C_2H_4 + CH_3$	2×10^{7}		0.0	11
6)	$C_3H_4 + M \rightleftharpoons C_3H_3 + H + M$	2×10^{17}		65	11
7)	$C_3H_4 + H \rightleftharpoons C_3H_3 + H_2$	5×10^{12}		1.5	11
8)	$C_3H_4 + CH_3 \rightleftharpoons C_3H_3 + CH_4$	2×10^{12}		7.7	11
9)	$C_3H_6 + O_2 \rightleftharpoons CH_3CHO + CH_2O$	9.8×10^{10}	0.5	0.0	12
10)	$C_3H_6 + OH \rightleftharpoons CH_3CHO + CH_3$	8.1×10^{10}	0.5	1.0	12
11)	$C_3H_6 + O \rightleftharpoons C_3H_4 + H_2O$	8.1×10^{11}	0.5	0.0	12
12)	$O + C_3H_6 \rightleftharpoons CH_2O + C_2H_4$	1×10^{13}		0.0	3
13)	$C_3H_4 + O_2 \rightleftharpoons CH_3CO + HCO$	1.0×10^{11}	0.5	0.0	12
14)	$C_3H_3 + H_2O \rightleftharpoons OH + C_3H_4$	2.2×10^{10}	0.5	40	14
15)	$C_3H_3 + OH \rightleftharpoons O + C_3H_4$	2.8×10^{9}	0.5	27.9	14
16)	$C_3H_2 + OH \rightleftharpoons C_3H_3 + O$	7.8×10^{9}	0.5	26.5	14
17)	$C_3H_2 + H_2O \rightleftharpoons C_3H_3 + OH$	2.6×10^{10}	0.5	38.7	14
18)	$C_3H_2 + O \rightleftharpoons C_2H_2 + CO$	3×10^{13}			est 10
19)	$C_3H_3 + OH \rightleftharpoons C_2HO + CH_3$	2.6×10^{10}	0.5	15.5	14
20)	$C_3H_2 + OH \rightleftharpoons C_2H + CH_2O$	3×10^{13}			est 10

Table III (continued)

(21)	$C_2H + O_2 \rightleftharpoons CO + CHO$	1×10^{13}		3.5	3
(22)	$CH_3CHO + OH \rightleftharpoons CH_3CO + H_2O$	6.5×10^{10}	0.5	4.0	12
(23)	$CH_3CHO + H \rightleftharpoons CH_3CO + H_2$	2.4×10^{12}	0.5	11.0	12
(24)	$CH_3CO + O \rightleftharpoons CH_3 + CO_2$	6.1×10^{11}	0.5	-	12
(25)	$CH_3CO + O_2 \rightleftharpoons CH_2OH + CO_2$	7.8×10^{10}	0.5	1.5	12
(26)	$CH_3CO + H \rightleftharpoons CH_3 + HCO$	2.2×10^{12}	0.5	5.4	12
(27)	$CH_2OH + H \rightleftharpoons CH_3 + OH$	2.2×10^{12}	0.5	10.7	12
(28)	$C_2H_4 + O \rightleftharpoons CH_3 + HCO$	3×10^{13}	-	-	12
(29)	$C_2H_4 + O \rightleftharpoons C_2H_2 + H_2O$	3×10^{13}	-	-	12
(30)	$C_2H_4 + OH \rightleftharpoons CH_3 + CH_2O$	1×10^{11}	0.5	7.1	12
(31)	$C_2H + O_2 \rightleftharpoons CH + CO_2$	1×10^{14}	-	23	12
(32)	$CH + O_2 \rightleftharpoons CO + OH$	8.1×10^{10}	0.5	-	12
(33)	$C_2H_2 + OH \rightleftharpoons C_2H + H_2O$	2.2×10^{14}	-	7	12
(34)	$C_2H_2 + O \rightleftharpoons C_2H + OH$	3.4×10^{15}	-0.64	18.7	12
(35)	$CH_2O + M \rightleftharpoons H_2 + CO$	2.1×10^{16}	-	35	12
(36)	$CH_2O + OH \rightleftharpoons HCO + H_2O$	2×10^{13}	-	-	12
(37)	$HCO + M \rightleftharpoons H + CO$	2×10^{12}	0.5	28.6	12
(38)	$O_2 + M \rightleftharpoons 2O$	2.6×10^{18}	-1.0	118	12
(39)	$CO + O_2 \rightleftharpoons O + CO_2$	1.6×10^{13}	-	41.1	12
(40)	$O + H_2 \rightleftharpoons OH + H$	1.8×10^{10}	1.0	8.9	12
(41)	$H_2 + O_2 \rightleftharpoons 2OH$	1.7×10^{13}	-	42.2	12

List of Figures

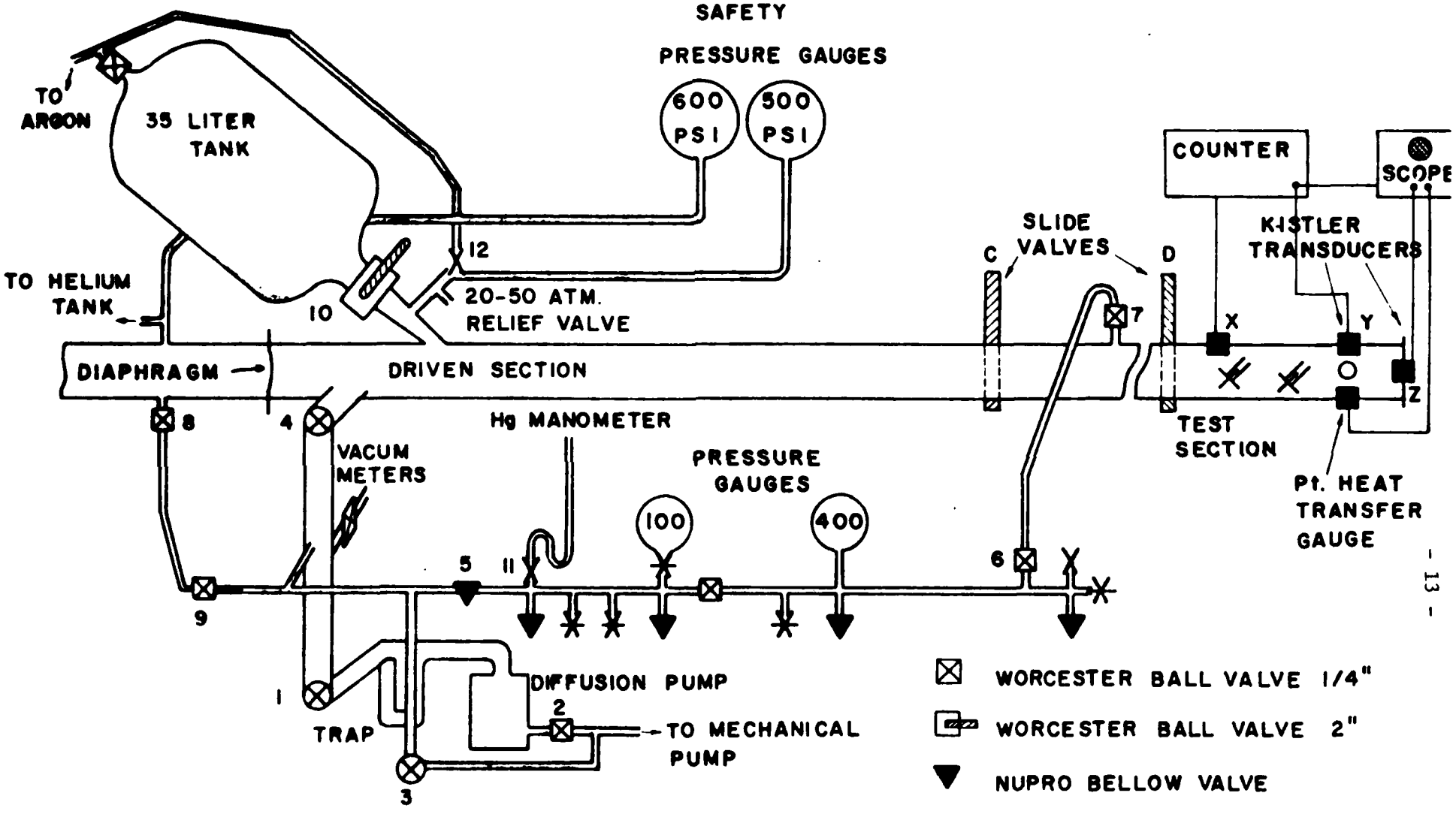


Fig. 1: Diagram of the Shock Tube.

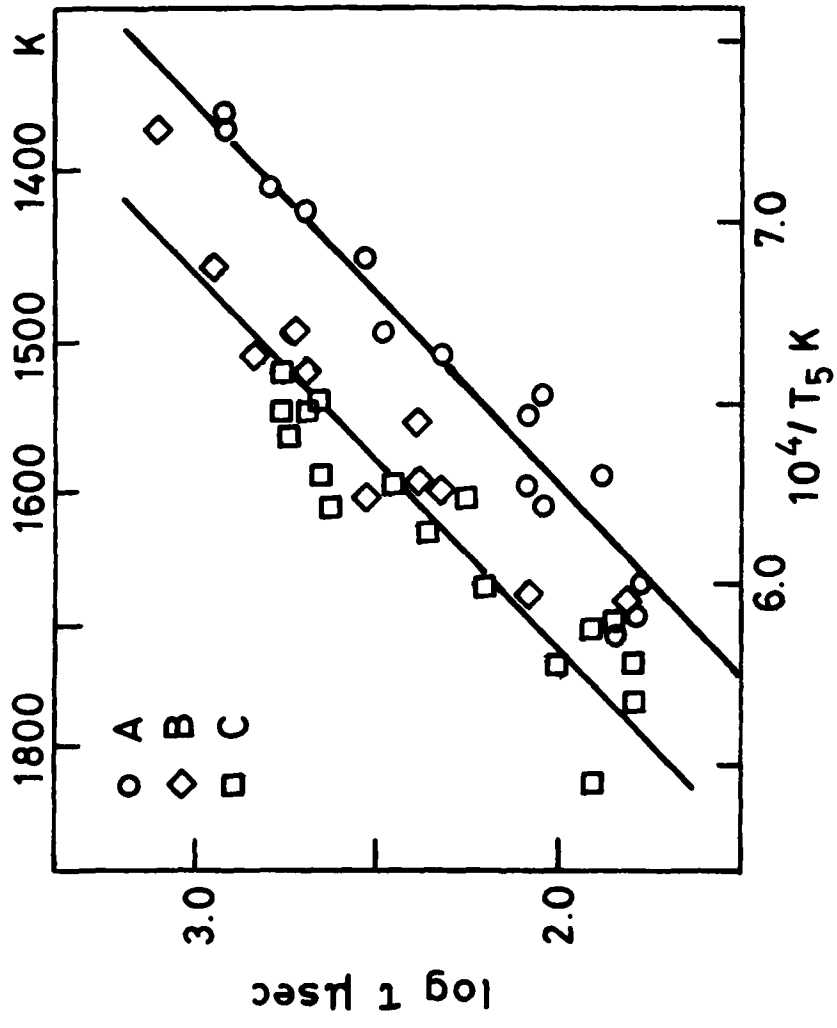


Fig. 2: A plot of log τ vs $1/T_5$ for mixtures A, B and C. Mixtures B and C show the argon power dependence over a concentration difference of 3. The spread shows a zero dependence.

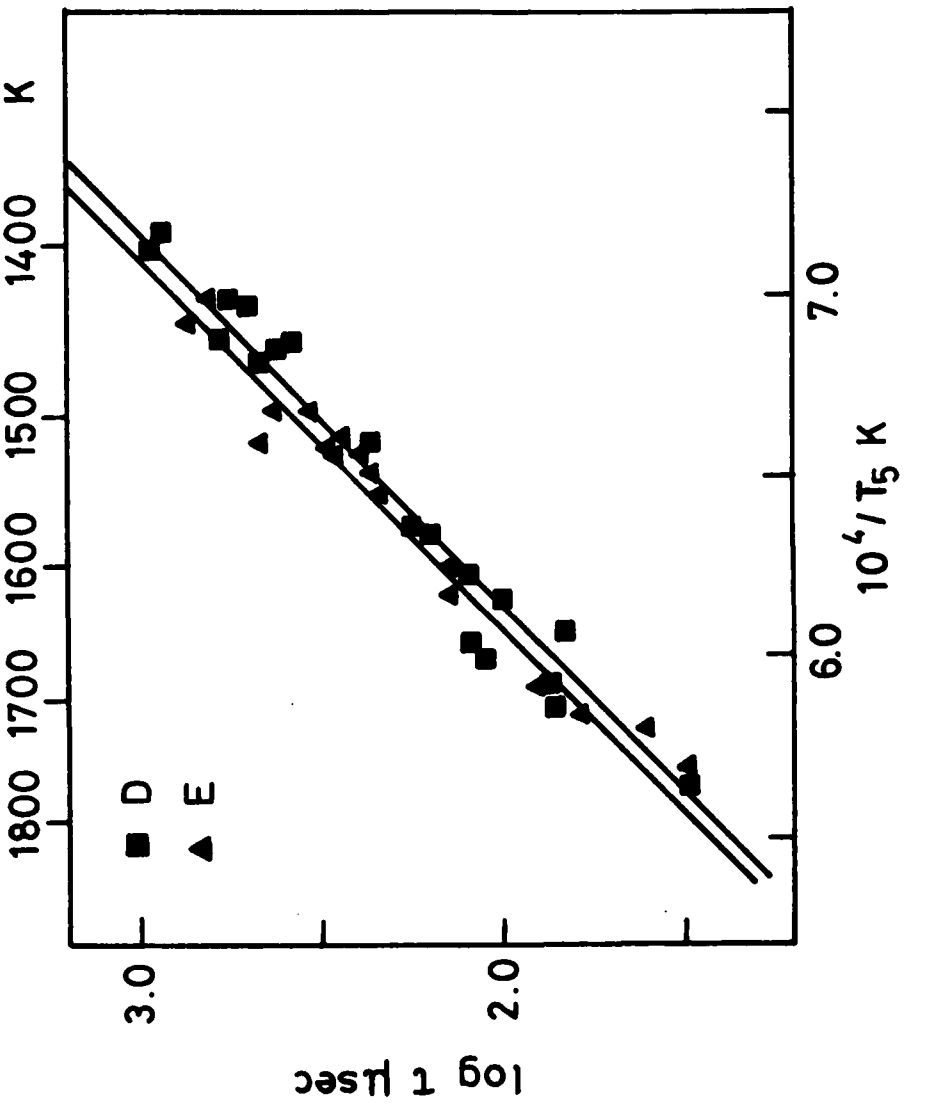


Fig. 3: A plot of log τ vs $1/T_5$ for mixtures D and E. The difference shows a negligible power dependence on the fuel.

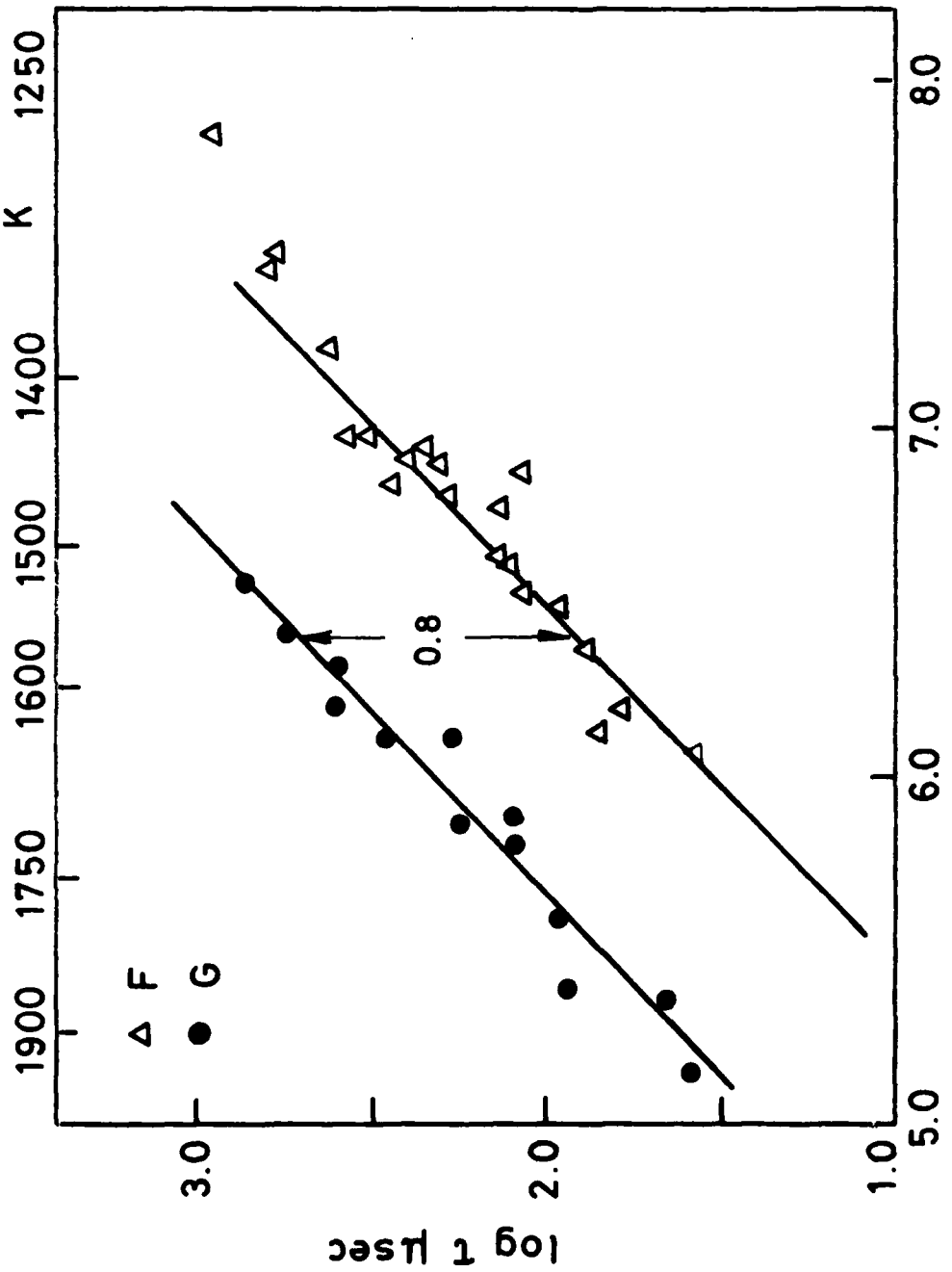


Fig. 4: A plot of log τ vs $1/T_5$ for mixtures F and G. The difference shows the power dependence of oxygen.

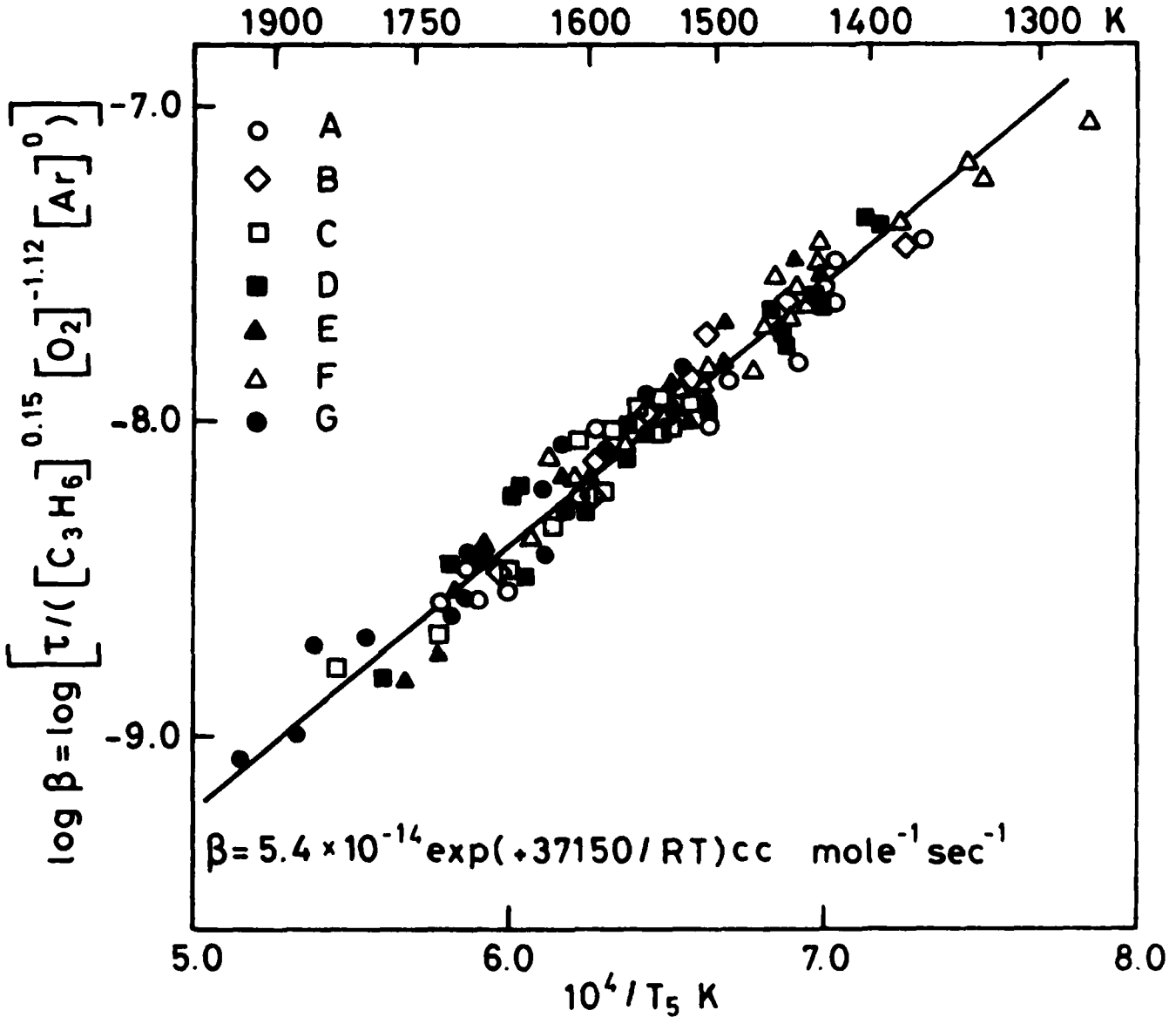
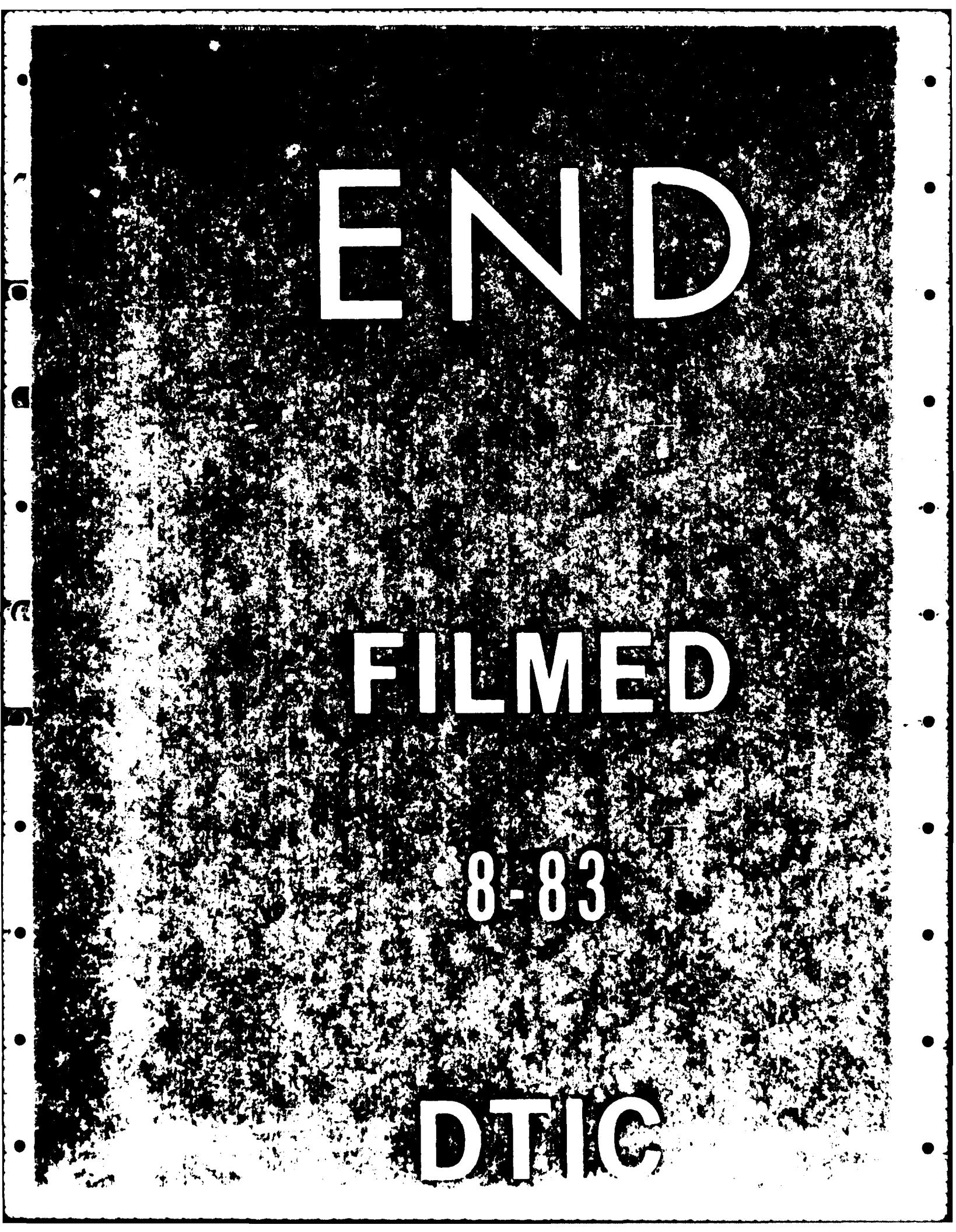


Fig. 5: A plot of log β vs $1/T_5$ for all the seven mixtures A to G. The maximum statistical spread allowed was 2σ.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